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| <p>(54) Title: MULTILAYER ADHESIVE FILM</p> <p>(57) Abstract</p> <p>A multiple layer adhesive film that exhibits high temperature characteristics, barrier properties, puncture resistance, and tear resistance includes at least three layers: a core layer disposed between and adhered, bonded or laminated to two outer adhesive layers. Each adhesive layer is formed from a polymer blend that includes at least an ethylene homopolymer and an ethylene copolymer having polar moieties. The core layer includes an olefinic polymer with a melting point that is greater than that of any adhesive layer polymer. More than one multiple layer adhesive film may be used in a thermoformable laminate structure that includes layers fabricated from other polymeric and non-polymeric materials.</p> <div data-bbox="621 1115 1317 1297" style="text-align: center;"> </div> |                  |  |

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## MULTILAYER ADHESIVE FILM

This invention relates to adhesive films.

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Thermoplastic adhesive films are frequently used to bond together various layers of automotive interior panels, such as headliners and visors. In addition to bonding together layers in an automotive interior panel, the thermoplastic adhesive films can be used to impart thermoformability to, or enhance thermoformability of, such laminates. In such cases, the adhesive film helps retain a desired shape of an automobile interior panel after the laminate has been subjected to thermoforming. This is particularly important in situations where the laminate is free of thermosettable resins and rigid foam. In these cases, the thermoplastic film should remain relatively stiff at temperatures in excess of those which may be generated in a closed vehicle on a sunny day, to prevent sagging of the automobile interior panel (e.g., headliner) under such conditions.

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The multipurpose monolayer adhesive films currently used in thermoformable laminates for automobile interior panels (e.g., headliners) tend to have excellent adhesive properties. Although the stiffness of such monolayer adhesive films is acceptable for many applications, it would be desirable to provide an adhesive film for use in thermoformable laminates for making automobile interior panels that has both excellent adhesive properties and improved stiffness to impart excellent anti-sag properties to automobile interior panels, even at highly elevated (abuse) temperatures.

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A problem with known multipurpose monolayer adhesive films, which are generally formulated to maximize adhesion, is that such films can tend to become torn or punctured during thermoforming of a laminate containing such films, especially if the thermoforming process subjects the laminate to a relatively deep draw. This can be a serious problem in those cases where the film is expected to act as a barrier to prevent a liquid thermosettable adhesive composition (e.g., polyurethane adhesive system) from leaking or bleeding through to, and damaging, a face fabric (e.g., the fabric facing on the interior side of an automobile headliner). It would be desirable to provide an adhesive film for use in thermoformable laminates for automobile interior panel applications which exhibits enhanced elongation, extensibility, and improved tear strength in addition to excellent adhesive properties like those of conventional multipurpose monolayer adhesive films and, desirably, improved stiffness (e.g., higher modulus).

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The multiple layer adhesive films of this invention have higher moduli than conventional monolayer adhesive films, and, therefore, impart improved sag resistance at elevated temperatures to laminated structures that are bonded together with these adhesive films. The multiple layer adhesive films also exhibit enhanced elongation,  
5 extensibility, and improved tear resistance, thus providing excellent barrier properties when used to bond together various layers of formed laminar articles of manufacture including automotive interior panels, such as headliners, other automotive applications, such as trunk liners, and any of a variety of non-automotive end uses. The formed laminar articles are preferably prepared via conventional thermoforming procedures that include use of a  
10 closed mold with temperatures in excess of 300° Fahrenheit (148.9°C) and pressures of from 10 psi (68.9 kilopascals (KPa)) up to and including 60 psi (413.7 KPa). The improvements and enhancements are relative to conventional multipurpose monolayer films.

The multiple layer adhesive films of this invention comprise a core layer  
15 having a first side and an opposing second side; a first outer adhesive layer bonded to the first side of the core layer; a second outer adhesive layers bonded to the second side of the core layer; the adhesive layers each comprising a polymer blend including an ethylene homopolymer and an ethylene copolymer having polar moieties; and the core layer comprising an olefinic polymer having a melting point greater than the ethylene  
20 homopolymer in each of the adhesive layers. Bonding preferably occurs by way of co-extrusion.

This invention also includes a method of preparing a formed laminar article of manufacture, the method comprising: providing a thermoformable laminar structure that includes at least one pair of two substrate layers and, disposed between the substrate  
25 layers, a multiple layer adhesive film as described in the immediately preceding paragraph; and subjecting the laminar structure to conditions of heat and pressure sufficient to convert the structure to the formed laminar article of manufacture. The formed article is suitably an automotive panel such as a headliner.

FIG. 1 is a schematic representation of a multiple layer adhesive film in  
30 accordance with the invention.

FIG. 2 is a schematic cross-sectional representation of an automotive headliner in which the multiple layer adhesive film is used to adhere a face fabric to other layers of the headliner..

FIG. 3 is a schematic cross-sectional representation of an automotive  
35 headliner in which the multiple layer adhesive films are used for bonding together various layers of the headliner.

A multiple layer adhesive film 10 is shown in FIG. 1. The multiple layer adhesive film includes a core layer 12, a first outer adhesive layer 13 laminated to a first side of the core layer 12, and a second outer adhesive layer 14 laminated to a second, opposite side of the core layer 12. Adhesive layers 13 and 14 are formulated to optimize  
5   adhesion at temperatures in excess of 115° centigrade (°C). In accordance with preferred embodiments, the adhesive layers are also formulated to provide chemical functionality to enhance adhesive characteristics and to bond skin layers (not shown in Fig. 1) to core layer 12. The multiple layer adhesive film is particularly well adapted for use in bonding various layers of a thermoformable laminate that is shaped or contoured under application of heat  
10   and pressure in a molding tool to form a contoured laminated article such as an automotive headliner. Adhesive layers 13 and 14 are formulated to melt at normal thermoforming processing temperatures (e.g., 115°C to 125°C), whereas the core does not melt at such normal thermoforming processing temperatures. Core layer 12 serves as a barrier because it has a higher melting point than adhesive layers 13 and 14 and remains solid at such  
15   thermoforming temperatures.

Adhesive layers 13 and 14 comprise a polymer blend including an ethylene homopolymer and an ethylene copolymer. The polymers in the polymer blend are preferably selected to have melting points of from 115°C to 125°C. The expression "ethylene homopolymer" encompasses homopolymers consisting essentially of ethylene  
20   monomer units as well as ethylene polymers containing minor amounts of monomer units other than ethylene provided that such polymers have the characteristics substantially similar to an ethylene homopolymer consisting essentially of ethylene monomer units. Suitable ethylene homopolymers include linear low density polyethylene (LLDPE) and low density polyethylene (LDPE). Preferred LDPE polymers are prepared using a constrained  
25   geometry metallocene catalyst (CGC), such as those disclosed in U.S. Patent No. 5,272,236. A commercially available and suitable LLDPE for use in the polymer blend is DOWLEX™ 2247A, which is available from The Dow Chemical Company. A preferred LDPE is ELITE™ 5110, an enhanced polyethylene resin produced using a CGC (commercially available from The Dow Chemical Company).

30       The ethylene copolymer included in the polymer blend of adhesive layers 13 and 14 contains, or has polymerized therein, polar moieties. Such ethylene copolymers are compatible with the ethylene homopolymer. The polar moieties of the ethylene copolymer provide a polar functionality that improves adhesion with other materials. For example, the polar functionality enhances adhesion with urethane adhesives commonly employed in  
35   thermoformable laminates that are used to make automotive interior panels such as

headliners. Such ethylene copolymers include ethylene/acrylic acid (EAA) copolymer resins and graft-modified ethylene polymers, such as maleic anhydride-modified (MAH-modified) ethylene polymers, especially MAH-modified LLDPE. An EAA copolymer suitable for use in the polymer blend is PRIMACOR<sup>TM</sup> 1430 (commercially available from The Dow Chemical Company). An anhydride-modified LLDPE that is suitable for use in the polymer blend is BYNEL<sup>®</sup> 4104 (commercially available from E. I. duPont de Nemours and Company). The polar moiety-containing ethylene copolymer can be used in nearly any amount. However, the amount is preferably from 20 percent to 35 percent by weight (wt percent), based on adhesive layer weight. Amounts below 20 wt percent have a diminished ability to enhance adhesion, and amounts above 35 wt percent generally do not provide sufficient additional benefit to justify the higher cost associated therewith.

The polymer blend of adhesive layers 13 and 14 preferably contains a minor amount of an anti-blocking concentrate, such as 15 wt percent silicon dioxide (SiO<sub>2</sub>) in 85 wt percent polyethylene (e.g. CN-734 available from Southwest Plastics), the amounts totaling 100 wt percent. The polymer blend also preferably contains a minor amount of a processing aid, such as AMPACET<sup>TM</sup> 10562, which is 3 wt percent VITON<sup>®</sup> A (a fluoroelastomer available from DuPont Dow Elastomers, L.L.C.) in 97 wt percent polyethylene, the percentages being based on processing aid weight and totaling 100 percent. If desired, the polymer blend of adhesive layers 13 and 14 may also contain other additives such as colorants, UV stabilizers, antioxidants and flame retardants.

The polymers of core layer 12 preferably have a melting point above 125°C and provide improved strength, improved puncture resistance, improved tear resistance, and improved anti-sag characteristics relative to polymers having a lower melting point. Olefinic polymers having a relatively high (greater than 20,000 psi (138 MPa), desirably greater than 25,000 psi (172 MPa), preferably less than or equal to 50,000 psi (345 MPa) although values in excess of 345 MPa may be suitable for some applications) modulus are preferred. Examples of preferred olefinic polymers for core layer 12 include propylene homopolymers, propylene copolymers, and high density polyethylene (HDPE). Core layer 12 preferably comprises a polymer blend comprises at least one of the above olefinic polymers and a second olefinic polymer that enhances at least one of compatibility and adhesion between the core and the adhesive layers. Examples of second olefinic polymers include anhydride-modified polyolefins and ultra low density polyethylene (ULDPE). The second olefinic polymer is preferably an anhydride-modified polypropylene when the olefinic polymer of the core layer is a propylene homopolymer or a propylene copolymer. A suitable commercially available anhydride-modified polypropylene is BYNEL<sup>®</sup> 50E571 (E. I.

du Pont de Nemours and Company). When the olefinic polymer of core layer 12 is HDPE, a preferred second olefinic polymer is an ULDPE such as ATTANE® 4201 (commercially available from The Dow Chemical Company). The relative amounts of olefinic polymer and second olefinic polymer in the polymer blend of the core layer are not particularly critical.

5 However, the amount of olefinic polymer in core layer 12 is preferably within a range of from 25 percent to 75 percent of the weight of the core layer, and the amount of second olefinic polymer is within a range of from 75 percent to 25 percent of the weight of the core layer. The amounts of olefinic polymer and second olefinic polymer are based upon core layer weight and total 100 percent. When the amount of olefinic polymer in the core layer is

10 below 25 wt percent, the resulting multiple layer adhesive film has diminished strength properties, diminished puncture resistance, diminished tear resistance, and diminished anti-sag characteristics relative to olefinic polymer amounts within the 25-75 wt percent range. When the amount of second olefinic polymer is below 25 wt percent, at least one of adhesion and compatibility of core layer 12 with adhesive layers 13 and 14 is diminished

15 relative to second olefinic polymer amounts within the 25-75 wt percent range.

The total thickness of multiple layer adhesive film 10 is typically from 1 mil (25 micrometers ( $\mu\text{m}$ )) to 4 mils (100  $\mu\text{m}$ ), and more typically about 2 mils (50  $\mu\text{m}$ ). Multiple layer adhesive films 10 are preferably prepared by blown film extrusion. During extrusion, core layer 12 thickness can vary by as much as 15 percent. In order to ensure that core

20 layer 12 has sufficient thickness (despite minor variations in thickness of extruded core layer 12) to achieve the desired strength, puncture resistance, tear resistance, and anti-sag characteristics desired, it preferably comprises from 30 percent to 60 percent of the total thickness of film 10.

The multiple layer adhesive films of this invention may be either cast or

25 extruded, but are preferably co-extruded. Co-extrusion appears to provide better interlayer contact and bonding than alternate techniques such as casting each layer separately and then forming a multiple layer film. The films preferably have a 2 percent secant modulus, as determined by ASTM D882, of at least 25,000 pounds per square inch (psi) (172.4 megapascals (MPa)) at 23°C. In the case of cast film, the modulus is the same in all

30 directions. For extruded films, the modulus is typically greater in the transverse direction than in the machine direction. Thus, for cast films a 2 percent secant modulus of at least 25,000 psi (172.4 MPa) at 23°C is preferred in any direction, and for extruded films a 2 percent secant modulus of at least 25,000 psi (172.4 MPa) in the machine direction is desired.

The multiple layer adhesive films may be used to prepare a variety of articles of manufacture including, without limitation, automotive interior panels. In particular, the multiple layer adhesive films of this invention may be positioned between substrate layers to form a thermoformable laminate, which may be subsequently molded under application of heat and pressure to form an automotive panel. Substrate layers may include, for example, foamed plastics, fabrics, fibrous mats, and random fibers impregnated with thermosettable adhesives.

A typical application for the multiple layer adhesive films of this invention is illustrated in FIG. 2. FIG. 2 shows a fragmentary cross section of an automotive headliner 15. Starting from one side that is designated as "roof side", headliner 15 comprises a scrim layer 16 that is fabricated from a material such as a non-woven polyester, a layer 17 comprised of random glass fibers impregnated with a polyurethane adhesive, a rigid polyurethane foam layer 18, a layer 19 comprised of random glass fibers impregnated with a polyurethane adhesive, a multiple layer adhesive film 20 (prepared as described above), and a face fabric 21. Face fabric 21 is adhered to layer 19 by film 20. The improved strength properties, improved puncture resistance, and improved tear resistance of multiple layer adhesive film 20 allows a laminate comprising layers 15-21 to be more deeply drawn (i.e., highly contoured) during thermoforming without film 20 puncturing or tearing and without allowing uncured urethane adhesive composition in layer 19 to leak or bleed through to face fabric 21. Thus, film 20 provides a highly effective barrier that prevents damage to face fabric 21 during thermoforming of a laminate comprising layers 16-21, even when thermoforming includes a high (4 to 5 inches (10.2 to 12.7 centimeters) of draw into a mold) degree of drawing or contouring of the laminate. Thermoforming preferably employs a closed mold, a temperature in excess of 300° Fahrenheit (148.9°C) and an applied pressure of from 10 psi (68.9 KPa) up to and including 60 psi (413.7 KPa).

FIG. 3 illustrates another application of the multiple layer adhesive films of this invention in an automotive headliner formed from a thermoformable laminate 30. Laminate 30 comprises, starting from one side designated as "roof side", a non-woven polyester scrim layer 32, a multiple layer adhesive film layer 33, a layer 34 comprised of random glass fibers (dry), a second multiple layer adhesive film layer 35, a flexible urethane foam layer 36, a third multiple layer adhesive film layer 37, a second dry random glass fiber layer 38, a fourth multiple layer adhesive film layer 39, and a face fabric layer 40. Multiple layer adhesive film layers 33, 35, 37 and 39 allow thermoformable laminate 30 to be transformed into a desired shape by application of heat and pressure in a molding tool, and to retain that shape, without using wet or uncured adhesives like a thermosettable urethane adhesive composition.



The following non-limiting examples illustrate the invention.

#### EXAMPLE 1

5                   A multiple layer adhesive film comprising a core layer and two adhesive layers, one on each side of the core layer, was prepared by blown film extrusion. During the blown film extrusion process, the inner set point temperature was 390°F (199°C), the core set point temperature was 420°F (216°C), the outer set point temperature was 410°F (210°C), the die temperature was 410°F (210°C), the frost line temperature was 229°F (109°C), and the air ring temperature was 50°F (10°C). Each adhesive layer was prepared from a blend that comprised 64 wt percent ELITE™ 5110 polyethylene, 30 wt percent BYNEL™ 4104 anhydride-modified LLDPE, and 6 wt percent CN-734 anti-block compound (15 percent SiO<sub>2</sub> in 85 percent polyethylene), all percentages other than components of the anti-block compound being based on polymer blend weight. Anti-block compound component weights were based on compound weight. The polymer blend for the core was 15   60 wt percent ATTANE™ 4201 ULDPE, and 40 wt percent ALATHON™ M6210 HDPE, both percentages being based on polymer blend weight. All polymer pellets were fed to a screw extruder of a blown film coextrusion line. The core comprised approximately 60 percent of the thickness of the extruded multiple layer adhesive film, and each of the adhesive layers 20   comprised approximately 20 percent of the thickness of the multiple layer adhesive film. The percentages totaled 100 percent. The extruded multiple layer adhesive film had a total thickness of 2.01 mils (50.25 µm).

                  The extruded multiple layer adhesive film had a machine direction (extruded direction) modulus of 35,708 psi (246.2 MPa) at room temperature (23°C) and a transverse 25   direction (perpendicular to the machine direction) modulus of 50,235 psi (346.4 MPa). Differential scanning calorimetry (DSC) was used to determine that the extruded multiple layer adhesive film had a melting point of 126°C.

#### EXAMPLE 2

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                  A multiple layer adhesive film was prepared via blown film extrusion using an inner set point temperature of 410°F (210°C), a core set point temperature of 430°F (221°C), an outer set point temperature of 410°F (210°C), a die temperature of 410°F (210°C), a frost line temperature of 236°F (113°C), and an air ring temperature of 55°F

(13°C). The polymer blends of the adhesive layers were the same as those of Example 1. The polymer blend of the core was comprised of 60 weight percent HIMONT HY 6100 polypropylene and 40 wt percent BYNEL™ 50E571 anhydride-modified polypropylene. The core comprised approximately 50 percent of the total thickness of the multiple layer adhesive film, and each of the adhesive layers comprised approximately 25 percent of the total thickness of the multiple layer adhesive film with the percentages totaling 100 percent). The multiple layer adhesive film had a total thickness of 1.94 mils (48.5 µm).

The multiple layer adhesive film had a machine direction modulus of 34,504 psi (237.9 MPa) at 23°C and a transverse direction modulus of 60,717 psi (418.6 MPa) at 23°C. DSC was used to determine that the multiple layer adhesive film had melting points at 123°C and at 156°C.

### EXAMPLE 3

A multiple layer adhesive film was prepared via blown film extrusion using the temperatures specified in Example 2. The polymer blend for each of the adhesive layers comprised 53 weight percent DOWLEX® 2247 A LLDPE, 40 wt percent PRIMACOR® 1430 EAA copolymer resin, 6 wt percent of its anti-blocking concentrate of Example 1, and 1 wt percent AMPACET™ 10562 process aid. The polymer blend of the core comprised 70 wt percent INSPIRE® C103-04 polypropylene copolymer and 30 wt percent of the anhydride-modified polypropylene of Example 2. The core layer comprised approximately 40 percent of the total thickness of the multiple layer adhesive film, and each of the adhesive layers comprised approximately 30 percent of the total thickness of the multiple layer adhesive film with the percentages totaling 100 percent. The composition of each layer also totaled 100 percent. The multiple layer adhesive film had a total thickness of 2.25 mils (56.25µm).

The multiple layer adhesive film had a machine direction 2 percent secant modulus of 57,500 psi (396.4 MPa) at room temperature (23°C) and a transverse direction 2 percent secant modulus of 56,000 psi (386.1 MPa) at room temperature (23°C). DSC showed that the multiple layer adhesive film had melt point peaks at 96.9°C, 123.7°C, and 164.9°C.

### EXAMPLE 4

A multiple layer adhesive film was prepared via cast film extrusion. The

polymer blend for each of two adhesive layers comprised 54 wt percent of the same LLDPE as in Example 3, 40 wt percent of the same EAA as in Example 3 copolymer resin, 5 wt CN-744 antiblocking concentrate (20 percent SiO<sub>2</sub> in 80 percent polyethylene, available from Southwest Plastics), and 1 wt percent of the same process aid as in Example 3. The  
5 polymer blend of the core was the same as in Example 3. The core layer comprised approximately 30 percent of the total thickness of the multiple layer adhesive film, and each of the adhesive layers comprised approximately 35 percent of the total thickness of the multiple layer adhesive film with the percentages totaling 100 percent. The multiple layer adhesive film had a total of 1.98 mils (1.98  $\mu$ m).

10 The multiple layer adhesive film had a machine direction 1 percent secant modulus of 40,750 psi (281.0 MPa) at room temperature (23°C) and a transverse direction 1 percent secant modulus of 44,390 psi (306.0 MPa) at room temperature (23°C). Differential scanning calorimetry showed that the multiple layer adhesive film had peaks at 101.0°C, 126.2°C and 165.5°C.

#### 15 EXAMPLE 5

Three automotive headliners utilizing thermoplastic adhesive films were prepared. The automotive headliners were identical except for the adhesive film used. The first  
20 headliner, used as a control, was prepared utilizing a commercially available multipurpose monolayer adhesive film (INTEGRAL™ 909 from The Dow Chemical Company) which is commonly employed in automotive headliners. The multiple layer adhesive films of Examples 1 and 3 were used respectively in the second and third automotive headliners. A sag test was performed on each of the headliners. The sag test involved placing the  
25 headliner in a simulated roof bucket and subjecting it to a series of environmental conditions, including exposure to high (typically 110-120°C) temperatures which may be encountered in a closed vehicle on a sunny day. At the conclusion of the environmental cycling, each headliner was measured for sag at the edges, and the second headliner (the one utilizing the adhesive film from Example 1) was tested for sag at the center. The  
30 testing was in conformance with a Ford Motor Company specification that requires a maximum of 13 millimeters (mm) of sag at the center of the headliner and a maximum of 3.5 millimeters of sag at the edge of the headliner. The results are set forth below.

| <u>ADHESIVE FILM USED</u> | <u>EDGE SAG</u> | <u>CENTER SAG</u> |
|---------------------------|-----------------|-------------------|
| Control (INTEGRAL™ 909)   | 15 mm           | not measured      |
| Film From Example 1       | 4.5 mm          | 1.3 mm            |
| Film From Example 3       | 3.0 mm          | not measured      |

5 The results of these sag tests indicate that the multiple layer adhesive films of this invention impart significantly improved anti-sag characteristics to automotive panels formed from laminates containing these films.

It will be apparent to those skilled in the art that various modifications to the preferred embodiment of the invention as described herein can be made without departing from the spirit or scope of the invention as defined by the appended claims.

WHAT IS CLAIMED IS:

1. A multiple layer adhesive film, comprising: a core layer having a first side and an opposing second side; a first outer adhesive layer bonded to the first side of the core layer; a second outer adhesive layer bonded to the second side of the core layer; the adhesive layers each comprising a polymer blend including an ethylene homopolymer and an ethylene copolymer having polar moieties; and the core layer comprising an olefinic polymer having a melting point greater than the ethylene homopolymer in each of the adhesive layers.
2. The film of Claim 1, wherein the ethylene homopolymer in each adhesive layer is selected from the group consisting of linear low density polyethylene, and low density polyethylene.
3. The film of Claim 1, wherein the olefinic polymer in the core layer is selected from the group consisting of a propylene homopolymer, a propylene copolymer or a high density polyethylene.
4. The film of Claim 1, wherein the ethylene copolymer having polar moieties is an ethylene/acrylic acid copolymer or a graft-modified ethylene polymer selected from the group consisting of anhydride-modified ethylene polymers and anhydride-modified linear low density polyethylenes.
5. The film of Claim 1, wherein the core layer further comprises a second polymer that enhances at least one of compatibility with and adhesion between the core layer and the adhesive layers, the second polymer being an anhydride-modified polyolefin or an ultra low density polyethylene.
6. The film of Claim 3, wherein the core layer further comprises a second polymer that enhances at least one of compatibility with and adhesion between the core layer and the adhesive layers, the second polymer being an anhydride-modified polyolefin or an ultra low density polyethylene.
7. The film of Claim 1, wherein the core layer consists essentially of a polymer blend selected from a blend of a) a propylene copolymer and an anhydride-modified polypropylene, b) a high density polyethylene and an ultra low density polyethylene, or c) a propylene homopolymer and an anhydride-modified polypropylene.
8. The film of Claim 1, wherein the olefinic polymer in the core layer comprises from 25 percent to 75 percent of the weight of the core layer, the ethylene copolymer in each adhesive layer comprises from 20 percent to 35 percent of the weight of the respective adhesive layers and the core comprises from 30 percent to 60 percent of the thickness of the adhesive film.

9. The film of Claim 1, wherein the olefinic polymer of the core has a melting point above 125°C, the polymers of each of the adhesive layers have a melting point at or below 125°C, and the film has a 2 percent secant modulus of at least 25,000 psi (172.4 MPa) at 23°C as determined using ASTM D882.

5 10. The film of Claim 1, wherein the ethylene homopolymer in each adhesive layer is a low density polyethylene prepared by constrained geometry catalysis.

11. The film of Claim 9, wherein the olefinic polymer of the core has a melting point of at least 130°C.

10 12. The film of Claim 11, wherein the multiple layer adhesive film has a machine direction modulus of at least 30,000 psi (206.8 MPa) at 23°C and a transverse direction modulus of at least 40,000 psi (206.8 MPa) at 23°C.

15 13. A method of preparing a formed laminar article of manufacture, the method comprising: providing a thermoformable laminar structure that includes at least one pair of two substrate layers and, disposed between the substrate layers, the multiple layer adhesive film of any of Claims 1-12; and subjecting the laminar structure to conditions of heat and pressure sufficient to convert the structure to the formed laminar article of manufacture.

20 14. The method of Claim 13, wherein the conditions of heat and pressure include a temperature in excess of 300° Fahrenheit (148.9°C) and a pressure, in a closed mold, of from 10 psi (68.9 kilopascals (KPa)) up to and including 60 psi (413.7 KPa).

15. A formed laminar article of manufacture prepared by the method of Claim 13, the formed article being an automotive panel selected from the group consisting of automotive interior panels and trunk liners.

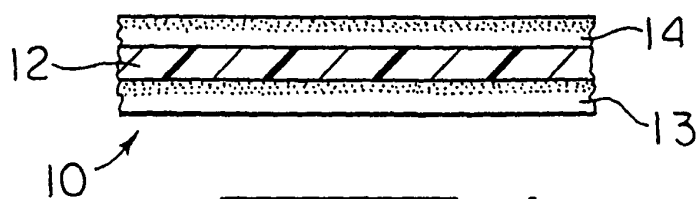


Fig. 1

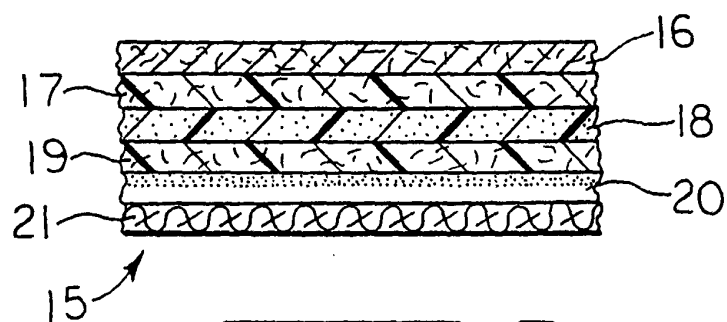


Fig. 2

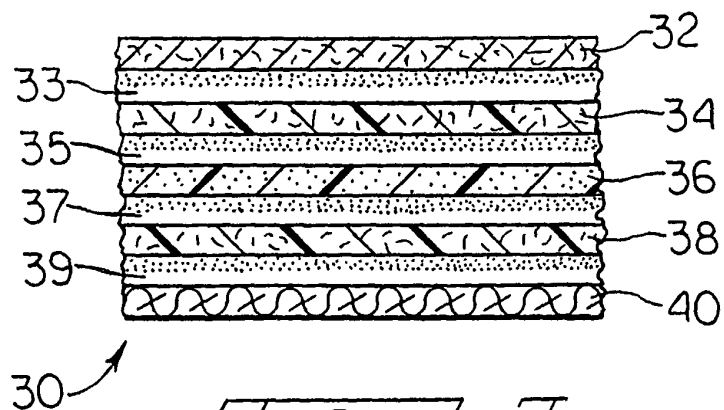


Fig. 3

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/12789

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C09J/02 B32B27/32

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C09J B32B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|-----------------------|
| A          | EP 0 713 765 A (WOLFF WALSRODE AG)<br>29 May 1996 (1996-05-29)<br>claims           | 1-15                  |
| A          | DE 33 30 185 A (CHEMPLEX CO)<br>23 February 1984 (1984-02-23)<br>claims            | 1-15                  |

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

8 October 1999

Date of mailing of the international search report

20/10/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Oudot, R



# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 99/12789

| Patent document<br>cited in search report | Publication<br>date | Patent family<br>member(s) | Publication<br>date |
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